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(54) Title: BLENDS OF POLYESTERS AND IMIDE ESTERS		

(57) Abstract

Plasticized polyesters containing 1 to 35 wt % of selected imide ester plasticizers. The plasticized polyesters have substantially reduced volatility and smoking, melt viscosities, melting points, and glass transition temperature as well as improved processing characteristics. These plasticized compositions are useful as adhesives in the form of powders, fibers, pellets, rods, melt blown webs, and flexible films as well as for use in extruded flexible shapes (e.g., tubing).

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BLEND OF POLYESTERS
AND IMIDE ESTERS

5 Technical Field

 This invention relates to new plasticized polyesters containing 1 to 35 wt % of selected imide ester plasticizers. The plasticized polyesters have substantially reduced volatility and smoking, melt viscosities, melting points, and glass transition temperature as well as improved processing characteristics. These plasticized compositions are useful as adhesives in the form of powders, fibers, pellets, rods, melt blown webs, and flexible films as well as for use in extruded flexible shapes (e.g., tubing).

Background of the Invention

 Many polyester polymers are useful as hot-melt adhesives for product assembly and as fabric adhesives. However, when the molecular weight of these polyester polymers is high enough to provide strong bonds, the melt viscosity of the polymers is so high that it is difficult, if not impossible, to apply these adhesives with conventional application equipment.

 Ester plasticizers are effective in modifying the properties of poly(vinyl chloride) and cellulose esters so that they may be used in the form of molded objects, tubing, film, sheeting, and the like. However, these ester plasticizers such as dioctyl phthalate, for example, are generally quite incompatible with polyesters. Other plasticizers, such as benzoate ester plasticizers, provide melt viscosities low enough that they can be applied with

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conventional application equipment for hot-melt adhesives. However, the polyester portion of these blends has a tendency to decrease in inherent viscosity (I.V.) when the blends are heated. U.S. Patent No. 4,172,824. U.S. Patent No. 4,094,721. U.S. Patent No. 3,186,961. U.S. Patent No. 2,044,612. Canadian Patent No. 919,190. British Patent No. 815,991, and U.S. Patent No. 4,340,625 are of interest.

10 Plasticizers used with polyester polymers generally encounter relatively high processing temperatures and shear conditions and the plasticizers tend to volatilize causing unacceptable conditions.

15 It has now been found that a limited number of imide esters are plasticizers for certain polyesters and that incorporation of these high-boiling plasticizers in these polyesters can significantly reduce the melting point and glass transition
20 temperatures of these polymers as well as to decrease the melt viscosity values of these blends. These high-boiling plasticizers show significantly reduced volatility and smoking when processed at high temperatures.

25 U.S. patent 4,172,824 discloses blends of certain poly(ethylene terephthalate) copolyesters containing adipic acid and 1,4-butanediol with selected benzoate ester plasticizers.

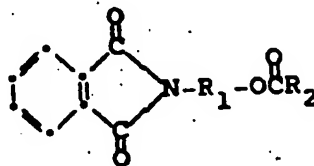
30 U.S. patent 4,450,250 discloses amorphous or crystallizable polyesters plasticized with mixed (aromatic and aliphatic) esters of phthalic anhydride and mixed phosphorus esters.

Disclosure of the Invention

35 According to the present invention, there is provided a plasticized polyester composition characterized as comprising

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- a) at least one amorphous copolyester or crystallizable copolyester having a melting temperature of less than 230°C. and a heat of fusion of less than 18 calories per gram, and
- b) a plasticizing amount of a compound of the formula



wherein R₁ is a linear or branched aliphatic hydrocarbon group having 2 to 20 carbon atoms or a cycloaliphatic or aromatic hydrocarbon group having 6 to 20 carbon atoms and R₂ is a linear or branched aliphatic hydrocarbon group having 1 to 20 carbon atoms, a cycloaliphatic hydrocarbon group having 5 to 20 carbon atoms or an aromatic hydrocarbon group having 6 to 20 carbon atoms.

The dicarboxylic acid component of the polyester may consist of aliphatic dicarboxylic acids having 4-12 carbon atoms, or alicyclic dicarboxylic acids, or aromatic dicarboxylic acids having 8-18 carbon atoms, or mixtures of two or more of these diacids. Examples of such dicarboxylic acids include succinic, glutaric, adipic, azelaic, sebacic, 1,12-dodecanedioic, 1,4-cyclohexanedicarboxylic, 1,3-cyclohexanedicarboxylic, terephthalic, isophthalic, and the like.

The glycol component of the polyester may consist of aliphatic diols having 2-12 carbon atoms and alicyclic diols having 6-18 carbon atoms, and

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selected poly(ethylene glycols) or mixtures of two or more of these diols. Examples of such diols include diethylene glycol, triethylene glycol, ethylene glycol, propylene glycol, 1,3-propanediol,

5 1,4-butanediol, 1,5-pentanediol, 1,6-hexanediol, 1,3-cyclohexanedimethanol, 1,4-cyclohexanedimethanol, and the like.

The polyesters are readily prepared using typical polycondensation techniques well known in the art. Typical polycondensation catalysts which may be
10 used include titanium alkoxides, dibutyl tin dilaurate, and combinations of zinc, manganese, or magnesium acetates or benzoates with antimony oxides or antimony triacetate.

15 Useful polymers may have I.V. values ranging from about 0.4 to about 1.2 with preferred polymers having I.V. values ranging from about 0.5 to 1.1.

The polymers are amorphous or crystallizable and generally have heats of fusion (ΔH_f) less than 12
20 calories/gram. The polymers generally have melting points measured by differential scanning calorimetry of 80°C to 230°C.

Operable polyesters include both amorphous and crystallizable polyesters. Preferred polyesters
25 include amorphous polyesters or relatively low melting polyesters with melting points up to about 230°C which have heats of fusion of less than about 18 calories per gram of polymer. Very high melting polymers such as poly(ethylene terephthalate) and
30 poly(1,4-cyclohexylenedimethylene terephthalate) generally are not operable in the practice of this invention because of the rather high temperatures (250°C and above) required for processing these blends.

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Some useful polyesters include poly(ethylene terephthalate) copolyesters modified with cyclohexanedimethanol or diethylene glycol, poly(tetramethylene terephthalate) modified with glutaric acid and diethylene glycol, poly(hexamethylene terephthalate) modified with glutaric acid and diethylene glycol, poly(1,4-cyclohexylenedimethylene 1,4-cyclohexanedicarboxylate) modified with trimellitic acid and poly(tetramethylene glycol), poly(hexamethylene terephthalate) modified with 1,4-butanediol, poly(hexamethylene terephthalate) modified with isophthalic acid and 1,4-butanediol, poly(tetramethylene terephthalate) modified with isophthalic acid, poly(1,4-cyclohexylenedimethylene terephthalate) modified with isophthalic acid, and the like.

The imide ester plasticizers of the compositions according to this invention are usually present in amounts of 1-35%, preferably 5-25% by weight of the compositions. In the structural formula given above for the imide ester, R_1 may contain 2 to about 20 carbon atoms and may be a linear or branched aliphatic hydrocarbon group; it may also contain cycloaliphatic or aromatic hydrocarbon groups but will generally have a methylene group adjacent to the ester and imide functions.

R_2 may contain 1 to 20 carbon atoms and may contain a linear or branched aliphatic hydrocarbon group, cycloaliphatic group, aromatic group, or alkyl-substituted cycloaliphatic or aromatic group. Such imide esters may be made using conventional techniques well known in the art.

The plasticizer may be incorporated into the polyester by heating the plasticizer and polymer at

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temperatures of about 75°C to about 250°C using mixing rolls, Banbury mills, extruders, and the like or the components may be dissolved in a mutual solvent followed by evaporation of the solvent.

5 The presence of the plasticizer provides significantly improved processing characteristics for the blends. For example, fibers and films may be extruded at temperatures lower than that required for the unmodified polyesters. The plasticized fibers
10 may be used as a binder for nonwoven polyester fabrics and the bonding is conducted at relatively low temperatures. Plasticized films are readily used for laminating fabrics or attaching labels and emblems at low bonding temperatures. Generally, the
15 plasticized films have better flexibility than the unmodified films and thus provide better hand for the bonded fabrics. Since the plasticizers are highly compatible with the polyesters, no exudation is noted on the surface of the films.

20 Plasticized polyester blends are readily reduced to powder by cryogenic grinding techniques using pinmill or hammermill grinders. These powders may be used on fusible interlining fabrics or used to bind nonwoven fabrics and bonding is achieved at
25 relatively low temperatures. In the binding of nonwoven fabrics, the plasticized powders may be applied to the nonwoven material from a water dispersion or by spraying the dry powders onto the fibers with an electrostatic spray gun. The dry
30 powders may also be applied from a scatter coating roll applicator such as a Schilling roll.

 The polyester blends are readily extruded into tubing and the tubing is much more flexible than the unmodified polyester. These flexible tubings are
35 quite easy to wind onto a core.

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The following examples are submitted for a better understanding of the invention.

Example 1

5 Polyester I pellets (0.5 g; Polyester I is a copolyester containing terephthalic acid, 69 mol % ethylene glycol, and 31 mol % 1,4-cyclohexanedimethanol; I.V. = 0.60, T_g = 73°C, amorphous; ΔH_f 0 cal/g; melt index at 190°C = . 9 g/10 minutes) are placed in a Pyrex test tube with 25 mL of 2,2-dimethyl-3-phthalimidopropyl 2-ethylhexanoate (boiling point 225-300°C at 3 mm) and the mixture is heated to 160°C with stirring. The polymer is dissolved readily and forms a clear solution. Similar results are obtained when 15 Polymer I is replaced with Polymer II. Polymer II is a copolyester containing 72 mol % terephthalic acid, 28 mol % glutaric acid, 55 mol % 1,4-butanediol, and 45 mol % diethylene glycol; (ΔH_f = 3.6 cal/g; 20 I.V. = 0.85; T_m = 110°C, T_g = 2°C).

Similarly good solubility results are obtained when 3-phthalimidopropyl 2-ethylhexanoate, 12-phthalimidododecyl 2-ethylhexanoate, 2,2-dimethyl-3-phthalimidopropyl butyrate, 2,2-dimethyl-25 3-phthalimidopropyl dodecanoate, 2,2-dimethyl-3-phthalimidopropyl p-dodecylbenzoate, 2-phthalimidoethyl p-decylbenzoate, [4-(phthalimidomethyl)cyclohexyl]methyl benzoate or 2,2-dimethyl-3-phthalimidopropyl 30 2,4,6-trimethylbenzoate are used.

Example 2 (Control)

The solubility test of Example 1 is repeated with Polyester I using dioctyl phthalate plasticizer

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at a temperature of 200°C. The polyester melts and forms a liquid layer in the bottom of the test tube but it does not dissolve or mix with the dioctyl phthalate. When cooled to 23°C, the polymer
5 solidifies and the plasticizer remains above the solid as a clear liquid. This example demonstrates that Polyester I is completely insoluble in a commonly employed plasticizer such as dioctyl phthalate in the 23°C to 200°C temperature range.

10

Example 3

Polyester I (47.5 g) is heated with 2,2-dimethyl-3-phthalimidopropyl 2-ethylhexanoate (2.5 g) in a Brabender Plastograph mixer at 190°C for
15 5 minutes in the melt phase to provide a blend containing 5 wt % plasticizer. The plasticized polyester has a melt index of 5.6 g/10 minutes at 190°C and a glass transition temperature of 71°C. The modulus of a 3 mil (0.08 mm) compression molded
20 film is 216,000 psi (15217 kg/cm²) by ASTM-882. For comparative purposes the unplasticized polyester has a melt flow rate of 2.9 g/10 minutes at 190°C and a glass transition temperature of 73°C. The modulus of
25 a 3 mil (0.08 mm) film of the unplasticized polyester is 264,000 psi (18,599 kg/cm²).

T-peel bonds (4 x 4 inches) (10 x 10 cm) are made on a Sentinel heat sealer with 3 mil (0.08 mm) compression molded films of unplasticized polyester
30 as well as with plasticized polyester using polyester/cotton twill fabric using five-second bonding time, 15 psi (1.06 kg/cm²) gage pressure and the temperature shown below. The bonds are cooled on a stone bench top. 1/2 inch (1.27 mm) is
35 trimmed from each side and three one-inch (2.54 cm)

T-peel bonds are cut from each specimen. Bonds are tested at 23°C on an Instron tester at a crosshead speed of 2 inches/minute (5.1 cm/minute) with the following results:

5

Bonding Temperatures		T-Peel Bond Strength, Plf (kg/linear cm)	
		Unplasticized	Plasticized
°F	(°C)		
10 284	(140)	1.3 (0.23)	6.2 (1.11)
320	(160)	4.4 (0.79)	11.5 (2.06)
356	(180)	13.6 (2.43)	19.8 (3.54)

Bonding temperatures above 400°F (204°C) scorch and degrade the fabric. This example illustrates that adding only 5% of the soluble plasticizer to the polyester lowers bonding temperature and increases bond strength. Bonds made with the plasticized polyester show excellent resistance to five cycles of commercial laundering and dry cleaning.

20

Example 4

The procedure of Example 3 is repeated except that Polyester I is blended with 25 wt % of 2,2-dimethyl-3-phthalimidopropyl 2-ethylhexanoate. The plasticized polyester has a melt index of 46.5 g/10 minutes at 190°C and a glass transition temperature of 30°C. The modulus of a 3 mil (0.08 mm) compression molded film by ASTM-882 is 28,000 psi (1973 kg/cm²).

30

Compared to the unplasticized polyester, the plasticized composition is easily extruded into film and tubing and injection molded into shaped articles at much lower pressures and shorter cycle times with no smoking or volatilization of the plasticizer.

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T-peel bonds 4 x 4 inches (10 x 10 cm) are made with 3-mil (0.08 mm) compression molded film of unplasticized polyester as well as with plasticized polyester samples using polyester/cotton twill fabric on a Sentinel heat sealer at the temperatures shown below [five-second bonding time; 15 psi (1.06 kg/cm²) gage bonding pressure]. Immediately after bonding, the bonds are quenched by placing the bonds on the stone bench top (temperature ~23°C) until the bonds have cooled to room temperature. One-half inch (1.27 cm) is trimmed from each side and three one-inch (2.54 cm) T-peel bonds are cut from each specimen. Bonds are tested at 23°C on an Instron tester at a crosshead speed of two inches/minute (5.1 cm/minute) with the following results:

Bonding Temperatures	T-Peel Bond Strength, Pli (kg/linear cm)			
	°F	(°C)	Unplasticized	Plasticized
20	284	(140)	1.3 (0.23)	26.0 (4.65)
	320	(160)	4.4 (0.79)	20.7 (3.70)
	356	(180)	13.6 (2.43)	10.8 (1.93)

The addition of 25 wt % 2,2-dimethyl-3-phthalimidopropyl 2-ethylhexanoate to Polyester I caused an increase in bond strength from 1.3 pli to 26.0 pli at a bonding temperature of 140°C.

Bonding temperature could be reduced even below 140°C with the plasticized polymer while the unplasticized polymer would not provide a bond at such low temperatures.

Similarly good results are obtained with [4-phthalimidomethyl)cyclohexyl]methyl benzoate.

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Example 5

Polyester I (4.25 pounds or 1.93 kg) and 0.75 pounds (0.34 kg) of 2,2-dimethyl-3-phthalimidopropyl 2-ethylhexanoate are melt blended in a 3/4 inch (1.9 cm) Brabender extruder at ~175°C and pelletized. The blend is cryogenically ground in a Mikro Pulverizer with liquid nitrogen using a 0.08-inch (2 mm) screen. After drying the powder at 40°C overnight under vacuum (25 inches of water; 635 kg/m²), 0.1 wt % calcium stearate is dry blended with the powder. Then the powder is classified by screening through 70 and 200 U.S. mesh screens by shaking for 15 minutes on a mechanical vibrator.

15 A polyester nonwoven web is formed by passing polyester staple fiber through a textile card machine to give a web weight of 2 ounces/yard² (47.5 gm/m²). Medium powder (70-200 mesh) of the plasticized polyester is fluidized with nitrogen and sprayed on the web with an electrostatic gun to give a uniform coating. The web is passed through a heating chamber of infrared heaters to fuse the adhesive powder and then through calendering rolls. The weight of adhesive in the web is 15 wt %. The web has a nice soft hand and is strong and uniformly bonded in both the machine and transverse directions. Bonding of a nonwoven web with 70-200 mesh powder of the unplasticized polyester in a similar manner gave a very weak web probably because of the high melt viscosity of the polyester and its inability to flow out.

Example 6

The procedure of Example 5 is repeated except that Polyester II is melt blended with 10 wt % of 2,2-dimethyl-3-phthalimidopropyl 2-ethylhexanoate.

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A nonwoven web bonded with medium powder (70-200 mesh) of this plasticized polyester has a nice soft hand and good strength in both the machine and transverse directions.

5

Example 7

- Polyester 1 is dissolved in methylene chloride with 10 wt % 2,2-dimethyl-3-phthalimidopropyl 2-ethylhexanoate (based on the weight of polyester).
- 10 The methylene chloride is evaporated and the resultant blend is granulated and dried at 40°C under vacuum (135 mm mercury) overnight. The blend is melt spun at 220°C using a 1.5 inch (3.8 cm) extruder equipped with a gear pump and a 45-hole spinnerette.
- 15 The fibers are air cooled and wound on a take-up roll. The filaments are subsequently drafted 3x at 70-100°C, crimped at 23°C with 12 crimps/inch (4.7 crimps/cm) in a stuffer box and cut into 1 1/2 inch staple fiber. A nonwoven web is prepared by mixing 20
- 20 wt % of this binder fiber with polyester staple fiber, passing the blend of fibers through a Shirley textile card machine and then through a bank of infrared heaters to fuse the binder fibers and bond the web. The nonwoven web has a nice soft hand and
- 25 good strength in both the machine and transverse direction.

Unmodified Polyester I is not spinnable into fibers at temperatures less than 300°C.

30 Example 8

Polyester I is melt blended with 15 wt % 2,2-dimethyl-3-phthalimidopropyl 2-ethylhexanoate in a 1 3/4 inch extruder at a melt temperature of ~225°C. The extruder blend is passed through a

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water cooling bath and pelletized. The pellets are dried under vacuum at 40°C overnight. The blend has a melt index of 16.0 g/10 minutes at 190°C and a T_g of ~48°C by DSC analysis. The blend is injection molded on a New Britain molding machine at a melt temperature of 230°C into a spiral mold. The plasticized polyester flowed 85 mm at a pressure of 700 psi (49.2 kg/cm²) compared to 56 mm for the unplasticized polyester.

10 The term "polyester" is used herein in a generic sense to include copolyesters. Also, esters of the acids rather than the acids themselves may be used in preparing the polyesters. For example, dimethyl terephthalate may be used in place of terephthalic acid if desired.

15 Although not required in the practice of this invention, small amounts of stabilizers, pigments, colorants, flow aids, anticaking agents, fluorescent agents or other additives may be used if desired.

20 Whenever the term "inherent viscosity" (I.V.) is used in this application, it will be understood to refer to viscosity determinations made at 25°C using 0.50 gram of polymer per 100 ml of a solvent composed of 60 wt % phenol and 40 wt % tetrachloroethane.

25 The "melting point" (T_m) of the polymers described in this application are readily obtained with a Differential Scanning Calorimeter.

The "heat of fusion" ΔH_f of polymers is the amount of heat absorbed when crystallizable polymers are melted. ΔH_f values are readily obtained using Differential Scanning Calorimeters (Perkin-Elmer). For example, one method for determining ΔH_f is described in Journal of Applied Polymer Science 20,1209 (1976). Measurement

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of ΔH_f is also described in duPont Thermal Analysis Bulletin No. 900-8 (1965). Qualitatively it is possible to compare the degree of crystallinity of polymers by comparing their ΔH_f values.

5 The strength of the bonds is determined by the so-called "Peel Test" based on the modification (i.e., three test specimens) of the ASTM "T-Peel Test" set forth on pages 63 and 64 of the 1964 edition of the BOOK OF ASTM STANDARDS, published by
10 the American Society for Testing Materials, and more specifically identified as Test Number D-1876-61-T.

Unless otherwise specified, all parts, percentages, ratios, etc., are by weight.

15 The invention has been described in detail with particular reference to preferred embodiments thereof, but it will be understood that variations and modifications can be effected within the spirit
20 and scope of the invention.

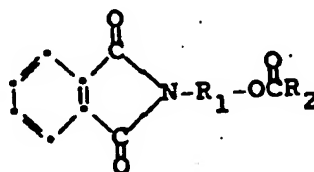
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Claims

1. A plasticized polyester composition characterized as comprising

- 5 a) at least one amorphous copolyester or crystallizable copolyester having a melting temperature of less than 230°C. and a heat of fusion of less than 18 calories per gram, and
- 10 b) a plasticizing amount of a compound of the formula



15 wherein R_1 is a linear or branched aliphatic hydrocarbon group having 2 to 20 carbon atoms or a cycloaliphatic or aromatic hydrocarbon group having 6 to 20 carbon atoms and R_2 is a linear or

20 branched aliphatic hydrocarbon group having 1 to 20 carbon atoms, a cycloaliphatic hydrocarbon group having 5 to 20 carbon atoms or an aromatic hydrocarbon group having 6 to 20 carbon atoms.

25

30 2. A composition according to Claim 1 characterized in that the copolyester contains repeat units from at least 40 mole % terephthalic acid, about 30 to 80 mole % ethylene glycol and about 70 to 20 mole % of another glycol selected from diethylene glycol and cyclohexanedimethanol.

35 3. A composition according to Claim 1 characterized in that the copolyester contains repeat units from terephthalic acid, glutaric acid, 1,4-butanediol and diethylene glycol.

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4. A composition according to Claim 1 characterized in that the copolyester contains repeat units from terephthalic acid, glutaric acid, 1,6-hexanediol and diethylene glycol.
5. A composition according to Claim 1 characterized in that the copolyester contains repeat units from 1,4-cyclohexanedicarboxylic acid, trimellitic acid, 1,4-cyclohexanedimethanol and poly(tetramethylene glycol).
6. A composition according to Claim 1 characterized in that the copolyester contains repeat units from terephthalic acid, isophthalic acid, 1,6-hexanediol and 1,4-butanediol.
7. A composition according to Claim 1 characterized in that the copolyester contains repeat units from terephthalic acid, isophthalic acid and 1,4-butanediol.
8. A composition according to Claim 1 characterized in that the copolyester contains repeat units from terephthalic acid, 1,4-butanediol and 1,6-hexanediol.
9. A composition according to Claim 1 characterized in that R_1 is a 2,2-dimethyltrimethylene group.

INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 86/02462

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) * According to International Patent Classification (IPC) or to both National Classification and IPC IPC ⁴ : C 08 K 5/34; C 08 L 67/02																	
II. FIELDS SEARCHED <div style="text-align: center; margin-top: 10px;">Minimum Documentation Searched ?</div> <table style="width: 100%; border: none;"> <tr> <td style="width: 30%; border: none;">Classification System</td> <td style="border: none;">Classification Symbols</td> </tr> <tr> <td style="border: none; padding-top: 10px;">IPC⁴</td> <td style="border: none; padding-top: 10px;">C 08 K</td> </tr> </table> <div style="text-align: center; margin-top: 10px; font-size: small;">Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched *</div>			Classification System	Classification Symbols	IPC ⁴	C 08 K											
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III. DOCUMENTS CONSIDERED TO BE RELEVANT* <table border="1" style="width: 100%; border-collapse: collapse;"> <thead> <tr> <th style="width: 10%; text-align: left; font-size: small;">Category *</th> <th style="width: 70%; text-align: left; font-size: small;">Citation of Document, ** with indication, where appropriate, of the relevant passages **</th> <th style="width: 20%; text-align: left; font-size: small;">Relevance to Claim No. **</th> </tr> </thead> <tbody> <tr> <td style="vertical-align: top; padding: 5px;">X</td> <td style="vertical-align: top; padding: 5px;">Research Disclosure, no. 253, May 1985 Kenneth Mason Publications Ltd (Empsworth, Hampshire, GB) "High solvating monomeric plasticizers", page 243, see example 1; column 1, lines 36-42</td> <td style="vertical-align: top; text-align: center; padding: 5px;">1-9</td> </tr> <tr> <td style="vertical-align: top; padding: 5px;">A</td> <td style="vertical-align: top; padding: 5px;">US, A, 3579363 (J.K. SEARS) 18 May 1971 see claim 1</td> <td style="vertical-align: top; text-align: center; padding: 5px;">1</td> </tr> <tr> <td style="vertical-align: top; padding: 5px;">A</td> <td style="vertical-align: top; padding: 5px;">US, A, 2684917 (W. GUNDEL et al.) 27 July 1954 see claim 1</td> <td style="vertical-align: top; text-align: center; padding: 5px;">1</td> </tr> <tr> <td colspan="3" style="text-align: center; padding: 10px;">-----</td> </tr> </tbody> </table>			Category *	Citation of Document, ** with indication, where appropriate, of the relevant passages **	Relevance to Claim No. **	X	Research Disclosure, no. 253, May 1985 Kenneth Mason Publications Ltd (Empsworth, Hampshire, GB) "High solvating monomeric plasticizers", page 243, see example 1; column 1, lines 36-42	1-9	A	US, A, 3579363 (J.K. SEARS) 18 May 1971 see claim 1	1	A	US, A, 2684917 (W. GUNDEL et al.) 27 July 1954 see claim 1	1	-----		
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A	US, A, 2684917 (W. GUNDEL et al.) 27 July 1954 see claim 1	1															

<div style="font-size: x-small;"> * Special categories of cited documents: ** "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "A" document member of the same patent family </div>																	
IV. CERTIFICATION <table style="width: 100%; border: none;"> <tr> <td style="width: 50%; border: none;"> Date of the Actual Completion of the International Search <div style="text-align: center; font-weight: bold;">23rd February 1987</div> </td> <td style="width: 50%; border: none;"> Date of Mailing of this International Search Report <div style="text-align: center; font-weight: bold;">27 MAR 1987</div> </td> </tr> <tr> <td style="width: 50%; border: none;"> International Searching Authority <div style="text-align: center; font-weight: bold;">EUROPEAN PATENT OFFICE</div> </td> <td style="width: 50%; border: none;"> Signature of Authorized Officer <div style="text-align: center;">M. VAN MOL </div> </td> </tr> </table>			Date of the Actual Completion of the International Search <div style="text-align: center; font-weight: bold;">23rd February 1987</div>	Date of Mailing of this International Search Report <div style="text-align: center; font-weight: bold;">27 MAR 1987</div>	International Searching Authority <div style="text-align: center; font-weight: bold;">EUROPEAN PATENT OFFICE</div>	Signature of Authorized Officer <div style="text-align: center;">M. VAN MOL </div>											
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ANNEX TO THE INTERNATIONAL SEARCH REPORT ON

INTERNATIONAL APPLICATION NO. PCT/US 86/02462 (SA 15281)

This Annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report. The members are as contained in the European Patent Office EDP file on 03/03/87

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Patent document cited in search report	Publication date	Patent family member(s)	Publicatio date
US-A- 3579363	18/05/71	None	
US-A- 2684917		None	

For more details about this annex :
see Official Journal of the European Patent Office, No. 12/82